

PROCEDURE FOR ABSORBED DOSE TO WATER DETERMINATION IN HIGH ENERGY PHOTON AND ELECTRON BEAMS BY FERROUS SULPHATE DOSIMETER AT INMRI-ENEA

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Abstract

Since 1998 the INMRI-ENEA has been using ferrous sulphate dosimeters to perform in-situ absorbed-dose-to-water calibration of radiotherapy clinical beams. The dosimeter consists of a ferrous sulphate solution in sealed glass ampoules of about 1 cm³ and with 0.5 mm wall thickness. The procedure adopted for the absorbed-dose-to-water determination and the recent improvements in the accuracy of the INMRI-ENEA ferrous sulphate dosimetric system are described. The most important improvements regard the standardization of the dosimeter shape and volume and the determination of the correction factor that accounts for the dosimeter non-water equivalence (glass wall and ferrous sulphate solution). The correction factors have been determined in photon and electron beams by Monte Carlo simulations using realistic spectra for the incident beams. The calculated values for the correction factor are reported as a function of the parameters TPR_{20/10} and R₅₀ for photon and electron beams, respectively. The new uncertainty budget for absorbed-dose-to-water measurement is reported and the major uncertainty components are discussed. At present the INMRI-ENEA ferrous sulphate dosimeter is a reference dosimeter used both for calibration and research activities.

Introduction

At the Istituto Nazionale di Metrologia delle Radiazioni Ionizzanti of ENEA (INMRI-ENEA) a dosimetric system based on ferrous sulphate solution was in the past years developed for performing in-situ calibration of radiotherapy electron beams characterized by very high dose per pulse (from 10 to 120 mGy per pulse). In these beams the D_w measurement by ionization chambers gives rise to some problems as the determination of the ion recombination correction by the traditional two-voltage method does not give coherent results [1]. On the contrary the response of ferrous sulphate dosimeter is independent of the dose per pulse up to value of 1 Gy per pulse [2]. At the INMRI-ENEA the ferrous sulphate dosimeter is presently used for a calibration service in terms of absorbed dose to water as well as for research activities. In this work the dosimetric system and the procedure adopted to make the D_w measurements traceable to the Italian Primary Standard of D_w are described. The procedure used for the D_w determination requires correction factors to take into account the non-water equivalence of the ferrous sulphate dosimeters (both dosimetric solution and dosimeter pyrex wall). The available literature data for these correction factors refer to dosimeters different from the INMRI-ENEA dosimeter for shape, dimension and wall thickness [3-5]. Then the correction factors have been specifically determined for the INMRI-ENEA dosimeters by Monte Carlo simulation based on the EGSnrc code [6]. Photon beams from Co-60 quality to 24 MV and electron beams with energy from 3 MeV to 20 MeV have been considered in the simulations.

Materials and methods

The INMRI-ENEA ferrous sulphate dosimeter consists of a sealed glass ampoule filled with a ferrous sulphate aqueous solution. The solution components are: 0.4 mol/L H₂SO₄, 10⁻³ mol/L Fe(NH₄)₂(SO₄)₂ (Mohr salt) and 10⁻³ mol/L NaCl. To improve the accuracy and stability of the solution, some H₂O₂ is used for oxidising the impurities contained in the acid and a small amount of KMnO₄ is then used to neutralise the excess of H₂O₂ [7]. The ferrous sulphate solution is prepared using Aristar concentrated sulphuric acid and ultra pure water provided by a Millipore Milli-Q 185 Plus system. Tap water is prepurified by a reverse osmosis treatment, which removes more than 95% of contaminants, and by an electro deionisation treatment, which further reduces the water ion content. The prepurified water is then exposed to UV light of wavelength 185 nm and 254 nm for a bactericidal action and for oxidising the remaining organic compounds. After a final purification and filter process the system provides water with a typical resistivity of about 18 MΩ cm at 25°C and total organic carbon less than 5 ppb. Ferrous sulphate solutions for reference dosimetry are prepared in 1 litre batches and each batch is used for periods not longer than three months.

The dosimeter ampoules have the following dimensions: inner diameter 7.8 mm, height 24 mm (referred to the top surface of the liquid in the ampoule) and pyrex wall thickness 0.5 mm therefore the volume of the dosimetric solution is about 1.1 cm³. The ampoule shape (fig 1) was specifically designed so to make it easier to fill and to seal the ampoule by flame without damaging the solution. The ampoule shape makes also possible irradiate the dosimeters in vertical beams without any air bubbles in the cylindrical part of the ampoule.

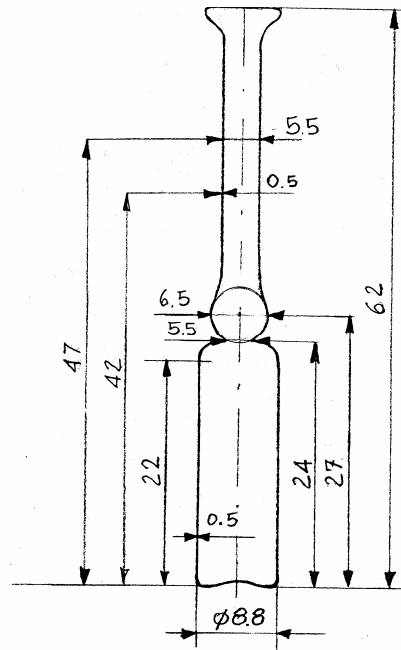


Figure 1. Schematic of the pyrex ampoule used for ferrous sulphate dosimeters at INMRI-ENEA.

The absorbance readings of ferrous sulphate solution are carried out by a Varian Cary 400 UV-VIS spectrophotometer. This instrument is provided with a Peltier thermostatic system allowing to perform absorbance measurements with the sample temperature stable within ± 0.1 °C. Absorbance readings are made at a wavelength of 304 nm with a bandwidth of 1 nm, using quartz micro cells having a pathlength of 10 mm, and an optical window 4 mm wide and 2 mm high. For each dosimeter two readings are performed using a couple of micro cells. After each absorbance reading the cells are cleaned with ultra-pure water and dried in a centrifuge.

The traceability of the D_w measurements is assured by calibrating each new batch of ferrous sulphate solution against the INMRI-ENEA absorbed-dose-to-water primary standard for the Co-60 gamma radiation [8]. Individually filled ampoules are irradiated at known D_w values in the Co-60 reference beam at a depth in water of 5 g cm⁻². The calibration coefficient, N_w , is determined as

$$N_w = D_w / \Delta A \quad (1)$$

where ΔA is the difference in absorbance between the irradiated and unirradiated ferrous sulphate solution. The absorbance readings are corrected for the influence of both the readout temperature, T_R , and the irradiation temperature, T_i [2, 9]. The reference temperature is 25 °C. To determine N_w and to check the linearity of the dosimeter response different values of D_w in the range from 40 Gy to 100 Gy are delivered to the dosimeters. The measured ΔA values are then fitted against D_w values to a straight line whose angular coefficient gives the N_w value.

The absorbed dose to water in high-energy photon and electron beams is determined as

$$[D_w]_Q = [\Delta A]_Q N_w F_Q \quad (2)$$

where the subscript Q refers to the beam quality and F_Q is a correction factor taking into account the energy dependence of the calibration coefficient.

As it is known, the absorbed dose to water as determined by ferrous sulphate dosimeter can be also expressed as

$$D_w = \frac{\Delta A}{\rho L \varepsilon G} P_{\text{FeSO}_4} P_{\text{wall}} \quad (3)$$

where ρ is the density of the ferrous sulphate solution (1.024 g cm^{-3}), L is the pathlength over which the optical density is read, ε is the molar extinction coefficient, G is the radiation yield of ferric ions, P_{FeSO_4} is a factor to convert the absorbed dose to ferrous sulphate solution in absorbed dose to water and P_{wall} is a correction factor taking into account the perturbation effects due to the dosimeter wall. Comparing equation (1) and (3) the calibration coefficient results to be

$$N_w = \frac{[P_{\text{FeSO}_4}]_{\text{Co-60}} [P_{\text{wall}}]_{\text{Co-60}}}{\rho L \varepsilon [G]_{\text{Co-60}}} \quad (4)$$

where the subscript Co-60 refers to the quality of the beam used to determine N_w . From equations (2), (3) and (4) the F_Q factor results

$$F_Q = \frac{[P_{\text{FeSO}_4}]_Q [P_{\text{wall}}]_Q}{[P_{\text{FeSO}_4}]_{\text{Co-60}} [P_{\text{wall}}]_{\text{Co-60}}} = \frac{[P]_Q}{[P]_{\text{Co-60}}} \quad (5)$$

where the symbol P denotes the total correction factor at a given beam quality. In obtaining equation (5) the G value has been considered independent of the beam quality. The G variation of about 1% reported in the Literature for the energy range considered in this work is taken into account assigning an uncertainty of 0.5% to the assumption of G energy independence [10].

The P_{FeSO_4} and P_{wall} factors were determined by Monte Carlo simulation as [4]

$$P_{\text{FeSO}_4} = D_w / D_{\text{FeSO}_4} \quad (6)$$

$$P_{\text{wall}} = D_{\text{FeSO}_4} / D_{\text{FeSO}_4, \text{pyrex}} \quad (7)$$

where $D_{\text{FeSO}_4, \text{pyrex}}$ is the absorbed dose in the ferrous sulphate solution inside the dosimeter with pyrex wall, D_{FeSO_4} is the absorbed dose in the ferrous sulphate solution inside the dosimeter with wall made of water and

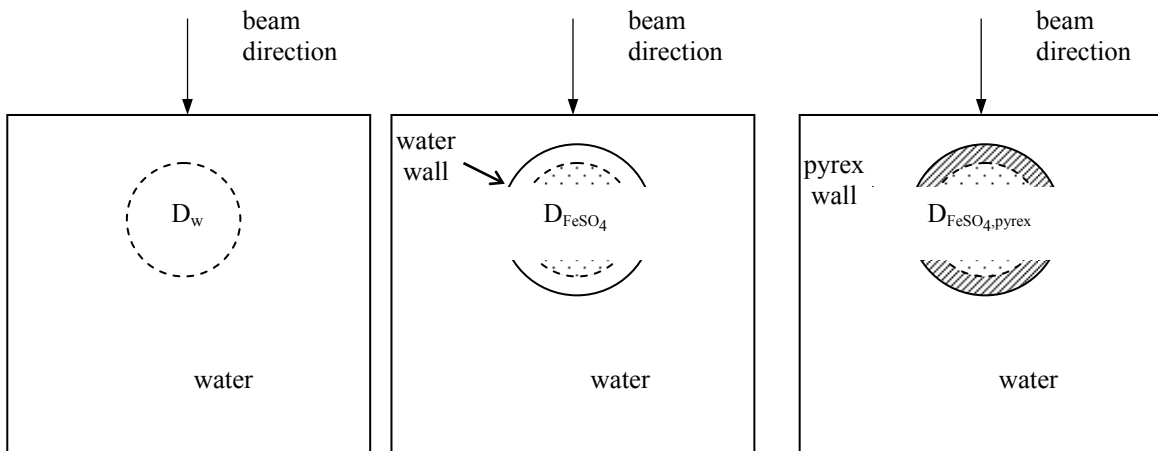


Figure 2. Schematic of the simulation geometry for P_{FeSO_4} and P_{wall} calculation according to the equations (6) and (7).

D_w is the absorbed dose to water in the water volume replacing the ferrous sulphate solution in absence of the dosimeter (Figure 2). The quantities D_w , D_{FeSO_4} and $D_{\text{FeSO}_4, \text{pyrex}}$ were obtained by scoring the energy deposited by the radiation beam in the region of interest (i.e. the volume occupied by the ferrous sulphate solution). The Monte Carlo calculation was performed by the EGSnrc/DOSRZnrc code which was modified to allow the scoring of the deposited energy in a cylindrical region placed in a cubic water phantom at various depths and having its axis parallel to the phantom surface (Figure 3). This geometry corresponds to the dosimeter irradiation conditions generally adopted in high-energy photon and electron beams. The calculation was carried out for electron beams with quality index R_{50} in the range from 1.5 g cm^{-2} to 8.5 g cm^{-2} , for photon beams with quality index $\text{TPR}_{20,10}$ in the range from 0.6 to 0.8 and for Co-60 gamma beam. The energy of the incident particles was sampled from the spectral distributions describing actual beams. As shown in table 1 electron beams produced by various types of accelerators have been considered. The energy spectra were taken from the Literature [11] or determined by simulating the actual beams with BEAMnrc code [12]. In particular the electron beams produced by the Hitesys NOVAC7[®] accelerator (an accelerator especially designed for intra-operative radiation treatment, IORT) were simulated. For comparison the correction factors were also determined for monoenergetic electrons with energy from 3 MeV to 24 MeV.

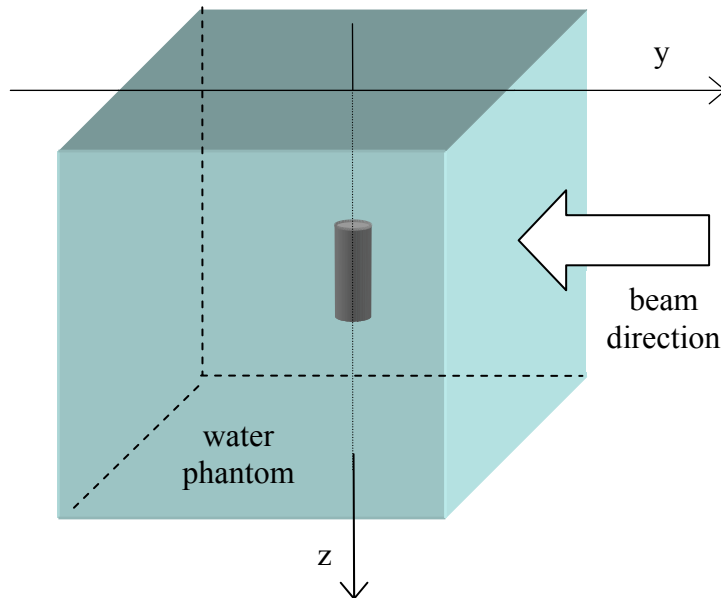


Figure 3. Schematic drawing (not to scale) of the dosimeter irradiation conditions considered in the Monte Carlo simulations.

The energy spectra for photon beams were taken from the Literature [13]. For the Co-60 quality the energy spectrum relevant to the INMRI-ENEA reference beam was used.

When available (i.e. for the NOVAC7[®] beams) the phase-space files describing the beam characteristics at phantom surface were used as input source for the modified EGSnrc/DOSRZnrc code. Otherwise parallel beams with direction perpendicular to the phantom surface and field size of 100 cm^2 were used.

For photon beams the depth in water of the scoring region was 5 cm for the beams with $\text{TPR}_{20/10}$ value less than 0.7 and 10 cm in the other cases. For electron beams the center of the scoring region was placed at the depth of the maximum dose, d_{max} . For each beam, the value of d_{max} was obtained from the simulated depth dose curve in water.

In the simulations electrons were followed down to 10 keV and photons down to 1 keV (ECUT = 0.521 MeV and PCUT = 0.001 MeV).

Table 1. Characteristics of electron beams considered for the Monte Carlo calculation of correction factors accounting for non-water equivalence of the INMRI-ENEA ferrous sulphate dosimeters. The values of the quality index, R_{50} , and the depth of the maximum dose, d_{\max} , were determined by the simulated depth dose curves in water.

Accelerator type	Nominal energy MeV	R_{50} g cm^{-2}	d_{\max} cm
Hitesys NOVAC7 [®]	9	2.85	1.28
	7	2.50	1.08
	5	2.12	0.88
	3	1.64	0.73
Varian Clinac 2100C	18	7.97	4.10
	15	6.50	3.35
	12	5.18	3.05
	9	4.02	2.05
	6	2.66	1.45
Philips SL75 20	17	6.96	4.13
	10	4.12	2.20
	5	2.08	1.15
Therac AECL 20	17	6.85	4.30
	6	2.18	1.35

Results

In figure 4 the P_Q values determined in electron beams at the depth in water of d_{\max} are reported as a function of R_{50} . The uncertainty bar reported in the figure (about 0.2%) is the statistical uncertainty of the Monte Carlo calculation. The results in figure 4 show that for R_{50} greater than about 2.4 g cm^{-2} the factor P_Q is weakly dependent on the electron energy distribution. The P_Q value obtained using the realistic spectral distributions ranges from 1.000 to 1.006 and the differences between P_Q values obtained for beams with similar R_{50} are less than 0.2%. Such differences are up to 0.5% when the results obtained for monoenergetic electrons are also considered.

For R_{50} values less than 2.4 g cm^{-2} the correction factor tends rapidly to increase with decreasing R_{50} and differences up to about 1% were found out between P_Q values for electron beams with nearly the same R_{50} value.

In the range of R_{50} considered in this work the P_{FeSO_4} factor resulted approximately independent of the electron energy. The average value of P_{FeSO_4} is 1.0044 ± 0.0010 therefore the variation of P_Q with electron energy is essentially due to the P_{wall} factor.

The dependence of the P_Q value on the angular distribution of the incident particles and on the depth of measurement in water is still under investigation. On the basis of preliminary results the effect of the angular distribution of the electron beam on P_Q value seems to be of a few tenths of per cent. Furthermore, the variation of P_Q value is estimated to be about 0.3% per mm in the region around d_{\max} where the percentage depth dose is higher than 98%.

In figure 5 the P_{FeSO_4} , P_{wall} and P_Q factors are reported for photon beams as a function of $\text{TPR}_{20/10}$. The P_{FeSO_4} is approximately constant in photon beams too. Its average value is 1.0027 ± 0.0011 . The correction accounting for the perturbation effects due to the pyrex wall was found to range from 0.994 to 0.989. The P_Q value decreases from 0.997 in the Co-60 beam to 0.991 in photon beams with $\text{TPR}_{20,10}$ of about 0.8.

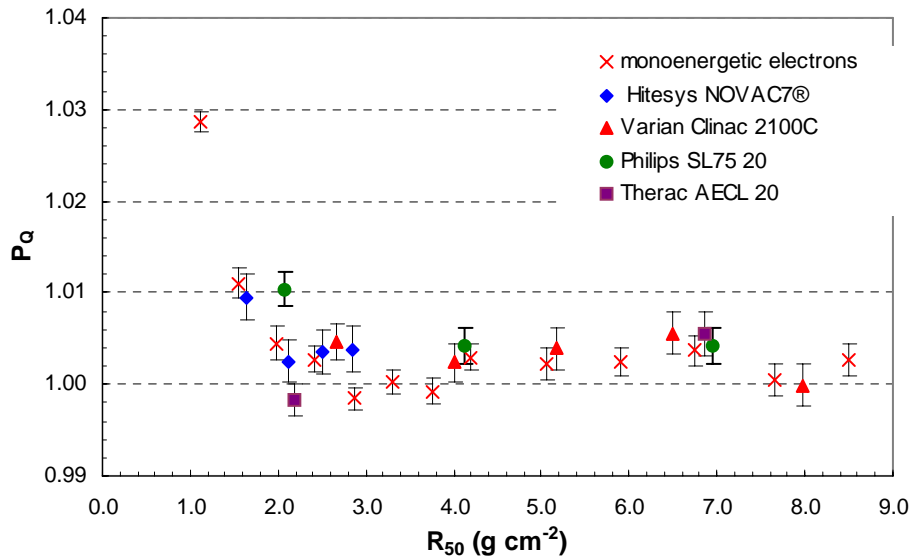


Figure 4. Correction factors, P_Q , taking into account the non-water equivalence of the INMRI-ENEA ferrous sulphate dosimeters in electron beams as a function of the R_{50} parameter. The factors were determined by Monte Carlo calculation for electron beams produced by accelerators of different type. The correction factors were calculated at the depth of maximum dose in water by sampling the energy for the incident electrons from the realistic spectral distributions. For comparison, the calculation was also carried out for monoenergetic electrons with energy in the range from 3 MeV to 24 MeV.

The linearity of the ferrous sulphate dosimeter response has been always verified. The correlation coefficient R^2 resulted equal to unit within 10^{-4} and the intercept of the linear fit resulted equal to zero within the uncertainty. In re-determining within the period of three months the calibration coefficient for the same batch of solution, a reproducibility of about 0.2% (1σ) was obtained. The calibration coefficient, as obtained at INMRI-ENEA in the period from 1998 to 2006, has a mean value of 278.7 Gy/ODU with a relative standard deviation of 0.7%. This value is consistent with the N_w value (276.7 Gy/ODU) obtained by equation (4) using $\epsilon G = 352 \cdot 10^{-6} \text{ m}^2 \text{ kg}^{-1} \text{ Gy}^{-1}$ (ICRU 64, 2001) and the value of P_{Co-60} determined in this work (0.997).

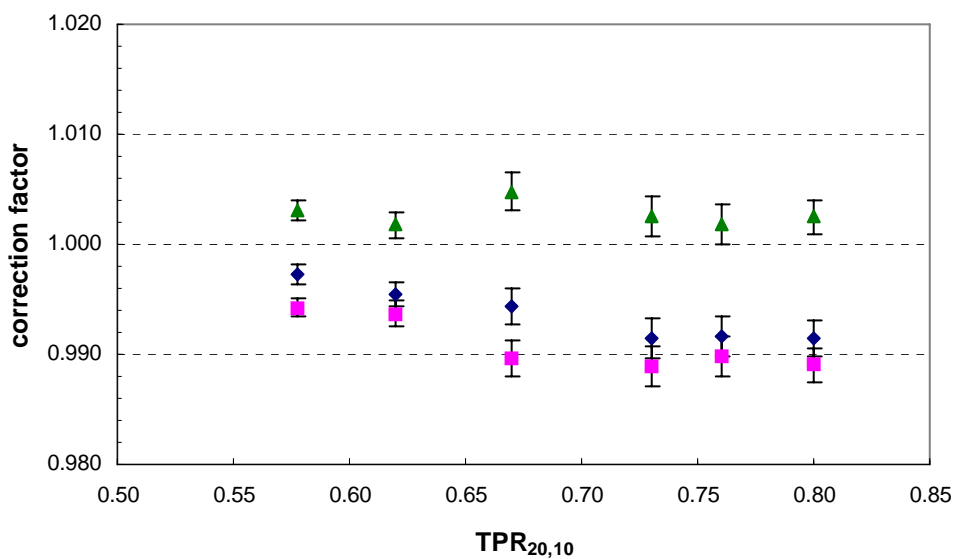


Figure 5. Correction factors P_{FeSO_4} (\blacktriangle), P_{wall} (\blacksquare) and P_Q (\blacklozenge) determined by Monte Carlo calculation for the INMRI-ENEA ferrous sulphate dosimeters in photon beams. The factors refer to the reference depth in water of 5 cm for the beams with $TPR_{20/10}$ value less than 0.7 and 10 cm for the other beams.

The uncertainty budget on D_w determination by INMRI-ENEA ferrous sulphate dosimeters in clinical beams is reported in table 2. The table refers to the D_w determination at the reference depth according to the current dosimetry protocols. The type A uncertainty component due to ΔA measurements refers to the mean value obtained by three dosimeters irradiated at the same dose. The type B uncertainty on F_Q factor (0.5 %) takes into account the uncertainty on modeling the experimental conditions in the Monte Carlo simulation. The value 0.5 % was obtained combining the following components: 0.3 % due to the depth of measurement (the typical agreement between simulated and measured percentage depth dose curves is ± 1 mm), 0.3 % due to the beam spectral distribution and 0.2 % due to the beam angular distribution. The combined standard uncertainty on D_w is 1.1%.

For the calibration service of radiotherapy beams a set of ferrous sulphate dosimeters is mailed to the radiotherapy centre where the dosimeters are irradiated in a water phantom. The INMRI-ENEA after the dosimeter readings returns to the customer a certificate reporting the absorbed dose to water delivered to each dosimeter. The combined standard uncertainty on the certified D_w values is 1.6% as each D_w value refers to a measurement performed by a single dosimeter.

Table 2. Combined relative standard uncertainty on D_w value obtained by the INMRI-ENEA ferrous sulphate dosimeters in high energy photon and electron beams.

Uncertainty source	Relative uncertainty	
	Type A (1 σ) %	Type B (1 σ) %
N_w determination		0.7
G energy independence		0.5
Stability of ferrous sulphate solution		0.2
ΔA measurement	0.2 ^(*)	0.3
T_i correction		0.1
T_R correction		0.2
F_Q factor	0.2	0.5
Quadratic sum	0.32	1.06
Combined standard uncertainty		1.1

^(*)Typical experimental standard deviation of ΔA obtained using three dosimeters irradiated at the same dose.

Conclusions

The INMRI-ENEA ferrous sulphate dosimetric system is currently used for calibrations of clinical beams in terms of D_w with particular regard to the electron beams with high dose per pulse. The procedure to perform such calibrations requires the knowledge of the correction factors accounting for the dosimeter non-water equivalence (both dosimetric solution and pyrex wall). These correction factors were calculated by Monte Carlo simulations based on the EGSnrc code using realistic energy spectra. In particular the correction factors have been determined at d_{max} for several electron beams produced by different types of accelerators and at the reference depth of 5 cm or 10 cm for photon beams with $TPR_{20/10}$ from 0.578 to 0.8. The combined standard uncertainty on D_w measurements is 1.1%.

The calibration service for electron beams with high dose per pulse is particularly requested for intra-operative radiotherapy accelerators in which ion recombination corrections cause problems for ionization chambers due to the high dose per pulse (above 10 mGy per pulse). Since 1998 more than 20 IORT accelerators have been calibrated by the ferrous sulphate in-situ dosimetry.

The ferrous sulphate dosimetry is currently used at INMRI-ENEA also for research activity as a method independent of the dose per pulse.

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